

Decolorization of C.I. Acid Yellow 23 solution by electrocoagulation process: Investigation of operational parameters and evaluation of specific electrical energy consumption (SEEC)

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Abstract

This study investigates the evaluation of specific electrical energy consumption (SEEC) and the influence of operating parameters on the color removal efficiency of a dye solution containing C.I. Acid Yellow 23 by electrocoagulation process. Firstly, the operational parameters including current density, initial dye concentration, initial pH and time of electrolysis were optimized. Then the effects of the conductivity, the interelectrode distance and the area of cross-section of the electrodes on specific electrical energy consumption (SEEC) were studied under the optimum conditions. Our results indicated that for a solution of 50 mg/l C.I. Acid Yellow 23, almost 98% color and 69% chemical oxygen demand (COD) were removed, when the pH was about 6, the time of electrolysis was 5 min and the current density was approximately 112.5 A/m². In addition, the results of our study revealed that when the conductivity and area of cross-section of the electrodes increased and interelectrode distance decreased, the cell voltage and specific electrical energy consumption would be decreased.

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Keywords: Electrocoagulation; Wastewater; Energy consumption; Current density; C.I. Acid Yellow 23

1. Introduction

Wastewaters generated by textile industries are known to contain considerable amounts of toxic aromatic dyes, especially azo dyes. It is well known that some azo dyes, and their degradation products such as aromatic amines are highly carcinogenic. The colored wastewater released into the ecosystem is a dramatic source of esthetic pollution and perturbation in the aquatic life. Conventional methods for dealing with textile wastewater consist of various combinations of biological, physical and chemical methods [1]. In recent years, investigations have been focused on the treatment of wastewaters using electrocoagulation (EC). The electrocoagulation has successfully been used for the treatment of wastewaters such as electroplating wastewater [2], laundry wastewater [3], latex particles [4], restaurant wastewater [5] and

slaughterhouse wastewater [6]. Meanwhile, EC process has been widely used in the removal of arsenic [7], phosphate [8], sulfide, sulfate and sulfite [9], boron [10], fluoride [11], nitrate [12] and chromate [13].

Treatments of wastewaters containing textile dyes have been studied by electrocoagulation method. The results of these studies show that COD, color, turbidity and dissolved solids at varying operating conditions are considerably removed [14–22]. In addition, it is clear that a technically efficient process must also be economically feasible with regard to its initial capital and operating costs, and practically applicable to the environmental problems. The economic aspect of the electrocoagulation (EC) process is not investigated well by the researches except a few of them [16]. Electrical energy consumption is a very important economical parameter in EC process like all other electrolytic processes.

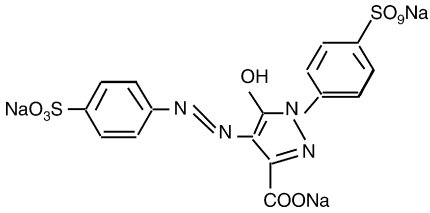
In this study the operational parameters such as the current density (CD), time of electrolysis, pH and initial dye concentration were optimized. Under the optimum conditions the effect of three operational parameters (conductivity (γ), interelectrode distance (d) and surface area (A)) on specific electrical energy

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Table 1
Structure and characteristics of C.I. Acid Yellow 23

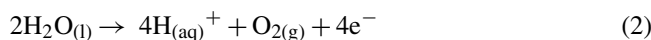
Dye	C.I. Acid Yellow 23
Structural	
λ_{\max} (nm)	425
Chemical class	Monoazo
C.I. number	19,140
M_w (g mol ⁻¹)	534.4

consumption was evaluated. C.I. Acid Yellow 23 (AY23) is soluble in water and belongs to acidic dyes group. C.I. Acid Yellow 23 is an azo dye present in thousands of textile (as a dye for wool and silk), foodstuff and pharmaceutical wastewaters. The chemical structure and other characteristics of this dye are shown in Table 1.

2. A brief description of EC

Electrocoagulation involves the generation of coagulants in situ by dissolving electrically either aluminum or iron ions in aluminum or iron electrodes, respectively. The metal ions generation takes place at the anode and the hydrogen gas is released from the cathode. The hydrogen gas would also help float the flocculated particles out of water. When a potential is applied from an external power source, the anode material undergoes oxidation, while the cathode will be subjected to reduction or reductive deposition of elemental metals. The electrochemical reactions with metal M as anode may be summarized as follows [18,23]:

- At the anode:



- At the cathode:



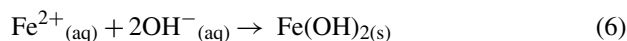
If iron or aluminum electrodes are used, the generated $Fe_{(aq)}^{3+}$ or $Al_{(aq)}^{3+}$ ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. For example, Al^{3+} ions on hydrolysis may generate $Al(H_2O)_6^{3+}$, $Al(H_2O)_5OH^{2+}$, $Al(H_2O)_4(OH)_2^{+}$ and the hydrolysis products may form many monomeric and polymeric species such as, $Al(OH)_2^{+}$, $Al_2(OH)_2^{4+}$, $Al(OH)_4^{-}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, $Al_{13}(OH)_{34}^{5+}$ in a wide pH range. Similarly, ferric ions generated by electrochemical oxidation of iron electrode may

form monomeric ions, $Fe(OH)_3$ and hydroxyl complexes, namely: $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^{+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$ and $Fe_2(H_2O)_6(OH)_4^{4+}$. Formation of these complexes depends strongly on solution pH [25]. Above pH > 9 $Al(OH)_4^{-}$ and $Fe(OH)_4^{-}$ are the dominant species. These hydroxides/polyhydroxides/polyhydroxy metallic compounds have strong affinity with dispersed/dissolved as well as the counter ions to cause coagulation/adsorption.

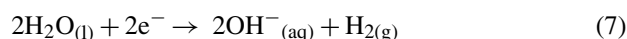
The gases evolved at the electrodes may impinge on and cause flotation of the coagulated materials [17]. Electrocoagulation of wastewater using iron electrodes takes place according to the following reactions: the following two mechanisms are involved for the production of $Fe(OH)_m$, where $m = 2$ or 3 [24].

• Mechanism 1

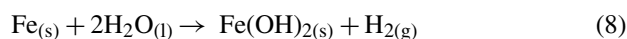
- Anodic reactions:



- Cathodic reaction:

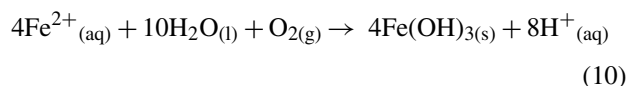


- Overall reaction:



• Mechanism 2

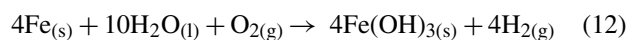
- Anodic reactions:



- Cathodic reaction:



- Overall reaction:



3. Materials and methods

The dye used in this project was purchased from Aldrich (Switzerland). Dye solution was prepared by dissolving dye in distilled water. The experimental set-up is shown in Fig. 1.

The conductivity of solutions was raised up and adjusted to different values by adding NaCl (Fluka, Switzerland). The conductivity measurement was carried out using a Philips conductivity meter (PW 9509, England). The pH of the solutions was measured by pH meter (Metrohm 654, Switzerland) and adjusted by adding NaOH or H_2SO_4 (Merck, Germany) solutions. Iron (ST 37-2) and aluminum plates were used as anode, and steel (grade 304) plates were used as cathode. Dimensions of electrodes were 40 mm × 50 mm × 1 mm and the distance between two electrodes in EC cell was 15 mm in all experiments. The electrodes were connected to a DC power supply

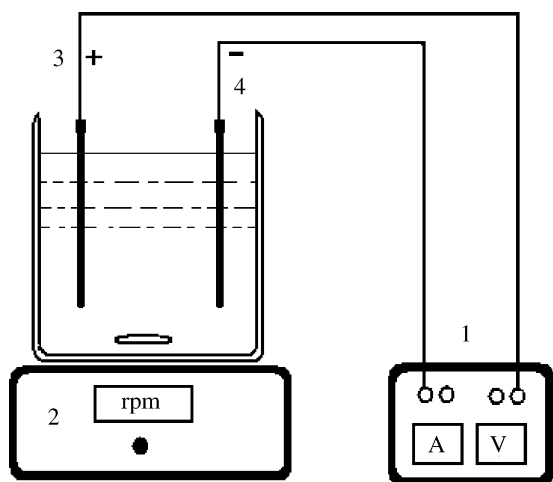


Fig. 1. An apparatus electrocoagulation cell: (1) digital DC power; (2) magnetic bar-stirrer; (3) anode; (4) cathode.

(ADAK PS808, Iran) with galvanostatic operational options for controlling the current density. The current density (CD) was calculated through the equation as follows:

$$CD = \frac{I(A)}{2S_{\text{electrode}}(m^2)} \quad (13)$$

where I is the current (A) and S is surface area of the electrode (m^2).

All the runs were performed at room temperature and stirring speed was 200 rpm. In each run, 250 ml of dye solution was placed into the electrolytic cell. The operation started when the current density was adjusted to a desired value. At the end of EC, the solution was filtered through 0.2 μm membrane filter (Schliecher & Schuell, Germany).

The reminiscent dye concentration was determined from its absorbent characteristics in the UV–Vis range (200–800 nm) with the calibration method using Beer–Lambert's law. A WPA lightwave (S 2000, England) spectrophotometer connected to a PC was implemented. The calculation of color removal efficiency (CR%) after electrocoagulation treatment was performed using this formula:

$$CR(\%) = \frac{C_0 - C}{C_0} \times 100 \quad (14)$$

where C_0 and C are concentrations of dye before and after electrocoagulation in mg/l, respectively. The chemical oxygen demand (COD) of dye solutions was measured based on the standard methods for examining water and wastewater (Ampoule method) [26].

4. Results and discussion

4.1. The effect of current density on the color removal efficiency

To investigate the effect of current density on the color removal efficiency, electrocoagulation process was carried out using various current densities by iron anode. Fig. 2 shows

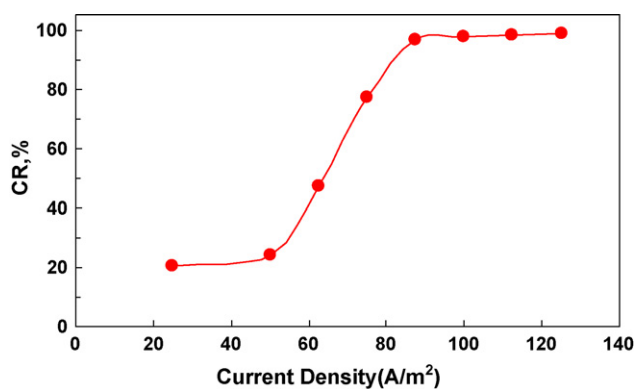


Fig. 2. Effect of current density on the color removal efficiency: $C_0 = 50$ mg/l; $t_{\text{EC}} = 5$ min; $\text{pH} = 5.35$; $\gamma = 16.5$ mS/cm; stirring speed = 200 rpm.

the color removal efficiency against current density applied to the iron electrodes in the EC process. The color removal efficiency increased to 98.60% at 125 A/m^2 from 20.66% at 25 A/m^2 . According to Faraday's law (Eq. (15)), when current density increases, the amount of ion produced on the electrodes increases. Therefore, there is an increase in floc production and hence an improvement in the color removal efficiency [18]. In other words, higher current density will generate significant amount of flocs, which in turn will trap the dye molecules and enhance the color removal efficiency.

$$\Delta M = \frac{MIt_{\text{EC}}}{nF} \quad (15)$$

where ΔM is the amount of iron dissolution (g); M the molecular weight of the iron (g/mol); n the number of electron moles; F is the Faraday constant ($F = 96,487$ C/mol). For a solution with a dye concentration of 50 mg/l, the optimum current density was 112.5 A/m^2 .

4.2. The effect of initial dye concentration on the color removal efficiency

The dye solution with different initial dye concentrations in the range of 20–200 mg/l was treated by EC in the optimized current density. The color removal efficiency was plotted against initial dye concentration at the electrocoagulation of 5 min (Fig. 3). As the results indicated, the color removal efficiency decreased with an increase in dye concentration. The presumed reason is deducible from Faraday's law (Eq. (15)). According to Faraday's law, constant amount of Fe^{2+} passed to the solution at the same current density and time for all dye concentrations. Consequently, the same amount of flocs would be produced in the solutions. As a result, the flocs produced at high dye concentration were insufficient to adsorb all of the dye molecules of the solution.

4.3. The effect of initial pH on the color removal efficiency

The EC process is highly dependent on the pH of the solution [24]. To examine its effect, the dye solutions were adjusted to

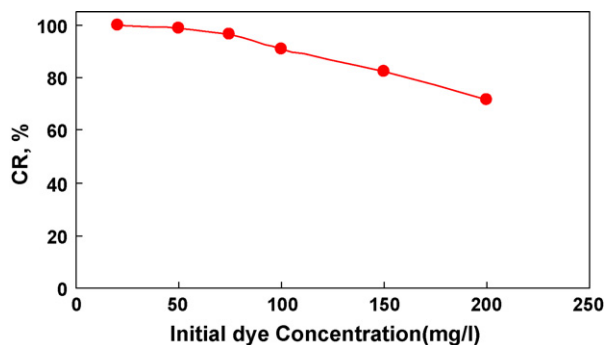


Fig. 3. Effect of initial concentration on the color removal efficiency: EC using iron electrodes, $CD = 112.5 \text{ A/m}^2$; $t_{Ec} = 5 \text{ min}$; $\text{pH} = 5.35$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

the desired pH for each experiment by adding sodium hydroxide or sulfuric acid solution. The color removal efficiency, as a function of pH, is shown in Fig. 4. It can be seen that the initial pH has a significant effect on the color removal efficiency especially at low pH. Close examination of the color removal efficiency versus pH clearly indicated that the color removal efficiency increased with pH and it remained unchanged between pH 5 and 9 and then on further increase of pH, the color removal efficiency decreased. Therefore, it can be concluded that at pH between 5 and 9, the majority of iron complexes (coagulants) are formed and it is the optimum pH for carrying out the electrocoagulation. There was minimum removal efficiency at the $\text{pH} < 2$. Since hydroxide ions, which were generated at the cathode, were neutralized by H^+ ions, sufficient amount of iron hydroxide complexes were not formed at this pH. At the $\text{pH} > 9$, color removal efficiency decreased as much as about 9%, because above this pH $\text{Fe}(\text{OH})_4^-$ is the dominant species. $\text{Fe}(\text{OH})_4^-$ is a dissolving species and it is unable to form flocs [10].

Since the initial pH value of C.I. Acid Yellow 23 solution was around 6, there is no need for the addition of chemicals to change the initial pH values. Generally, the medium pH changes during the process is also observed by other investigators [17,22]. This change depends on the type of electrode material and on the initial pH.

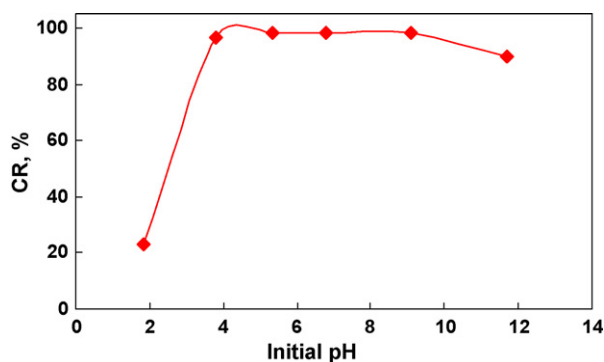


Fig. 4. Effect of initial pH on the color removal efficiency: EC using iron electrodes, $CD = 112.5 \text{ A/m}^2$; $t_{Ec} = 5 \text{ min}$; $C_0 = 50 \text{ mg/l}$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

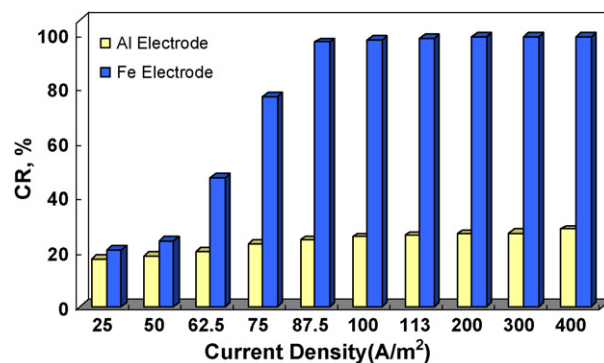


Fig. 5. Effect of electrode materials on the color removal efficiency: $C_0 = 50 \text{ mg/l}$; $t_{Ec} = 5 \text{ min}$; $\text{pH} = 5.35$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

4.4. The effect of electrode materials on the color removal efficiency

In any electrochemical process, electrode material has significant effect on the treatment efficiency. Therefore, appropriate selection of the electrode material is important. The electrode material for treatment of water and wastewater should also be non-toxic to human health and environment. Iron and aluminum were chosen as electrode material because these are non-toxic and easily available. To investigate the effect of electrode materials on the color removal efficiency, electrocoagulation process was carried out using iron and aluminum electrodes. Fig. 5 shows the color removal efficiency against current density applied to the electrodes in the EC process. As it is seen, iron electrode was more effective than aluminum electrode and as a result of increasing current density 16 times only about 11% additional color removal efficiency was achieved with aluminum electrodes. The plausible reason for lower color removal efficiency by aluminum in comparison with iron could be explained considering the insufficient ability of hydrous aluminum oxide to adsorb Acid Yellow 23 molecules. Also some investigators have reported that the adsorption capacity of hydrous aluminum oxide for dye molecules is much lower than that of the hydrous ferric oxide [7].

4.5. The effect of time of electrolysis on the color removal efficiency

During electrolysis, the positive electrode undergoes anodic reactions while cathodic reactions occur on the negative electrode. The released ions neutralize the particle charges and thereby initiate coagulation. The color removal efficiency depends directly on the concentration of ions produced by the electrodes. When the electrolysis period increases, the concentration of ions and their hydroxide flocs increases. Accordingly, as shown in Fig. 6, an increase in the time of electrolysis from 2 to 6 min yields an increase in the color removal efficiency from 15.53% to 98.98%. For a solution having a dye concentration of 50 mg/l and a treatment unit with current density of approximately 112.5 A/m^2 , the optimum time for electrolysis was 5 min.

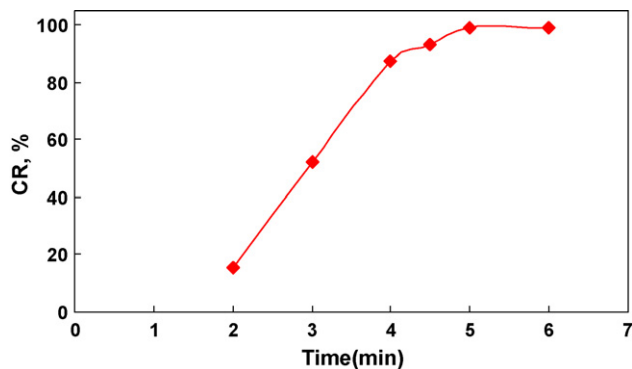


Fig. 6. Effect of electrolysis time on the color removal efficiency: EC using iron electrodes, $CD = 112.5 \text{ A/m}^2$; $C_0 = 50 \text{ mg/l}$; $\text{pH} = 5.35$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

4.6. The evaluation of specific electrical energy consumption (SEEC)

The current efficiency (φ) of EC process was calculated by Eq. (16). This calculation was based on the comparison of experimental weight loss of iron electrodes (ΔM_{exp}) during EC process with theoretical amount of iron dissolution (ΔM_{theo}) in line with the Faraday's law (Eq. (15)) [25]:

$$\varphi = \frac{\Delta M_{\text{exp}}}{\Delta M_{\text{theo}}} \times 100 \quad (16)$$

As $\text{Fe}(\text{OH})_{2(s)}$ is supposed to be the species formed at EC process by iron electrodes, the number of electron moles in dissolution reaction is equal to 2 (Eq. (5)). The specific electrical energy consumption (SEEC) was calculated as a function of iron electrodes weight consumption during EC in kWh/kg Fe using the following equation:

$$\text{SEEC} = \frac{nFU}{3.6 \times 10^3 M\varphi} \quad (17)$$

It is well known that the overpotential caused by solution resistance (IR-drop) has a significant effect on cell voltages (U) that depends on the distance (d in cm) between the electrodes, surface area (A in m^2) of the cathode and specific conductivity of the solution (γ in mS/m) and the current (I in A) [25].

4.6.1. The effect of conductivity on color removal efficiency and SEEC

To evaluate the effect of conductivity on color removal efficiency and specific electrical energy consumption, a number of experiments were performed. The conductivity of the solution was adjusted to the desired levels by adding an appropriate amount of NaCl to the dye solutions. Fig. 7 shows the effect of dye solution conductivity on the performance of the EC process. As it is seen, the color removal efficiency remains almost unchanged in the conductivity range of 2–26 mS/cm , whereas with the conductivity increasing, the specific electrical energy consumption (SEEC) was considerably (almost 68%) reduced. It is clear that the cell voltage decreased with an increase in the conductivity at constant current density. When the conductivity of solution increases, IR-drop decreases, so the necessary

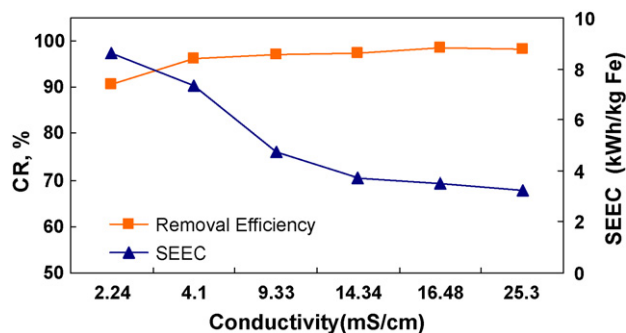


Fig. 7. Effect of solution conductivity on the color removal efficiency and SEEC: EC using iron electrodes, $CD = 112.5 \text{ A/m}^2$; $C_0 = 50 \text{ mg/l}$; $t_{\text{EC}} = 5 \text{ min}$; $\text{pH} = 5.35$; stirring speed = 200 rpm.

voltage to reach on optimum current density will be diminished, and consequently the consumed electrical energy is decreased.

4.6.2. The effect of interelectrode distance on color removal efficiency and SEEC

The effect of interelectrode distance on the removal efficiency and SEEC was examined at 0.5–3 cm. The effect of interelectrode distance on the color removal efficiency is shown in Fig. 8. When the interelectrode distance increases, the color removal efficiency increases slightly. Our results also showed that SEEC increased with an increase in the interelectrode distance (Fig. 8). The reason of this observation is thought to be the fact that when interelectrode distance increases, the ions produced would move more slowly, so IR-drop would increase. Hence, the voltage and SEEC for obtaining the optimum current density would increase.

4.6.3. The effect of area of cross-section of the electrodes on color removal efficiency and SEEC

The effect of area of cross-section of the electrodes on the removal efficiency and SEEC was evaluated at 5–30 cm^2 . Our results showed that SEEC decreased by increasing the area of cross-section of the electrodes, whereas the color removal efficiency remained almost unchanged (Fig. 9). When the area cross-section of the electrodes increases, IR-drop decreases, and thus the cell voltage and the electrical energy consumed are decreased.

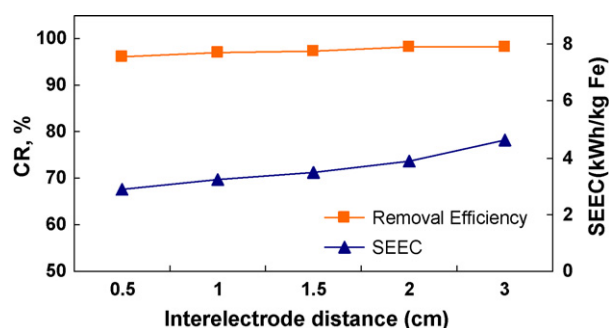


Fig. 8. Effect of interelectrode distance on the color removal efficiency and SEEC: EC using iron electrodes, $CD = 112.5 \text{ A/m}^2$; $C_0 = 50 \text{ mg/l}$; $t_{\text{EC}} = 5 \text{ min}$; $\text{pH} = 5.35$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

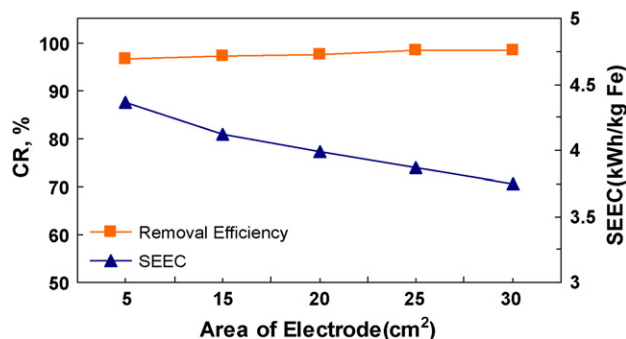


Fig. 9. Effect of area of cross-section of the electrodes on the color removal efficiency and SEEC: $CD = 112.5 \text{ A/m}^2$; $C_0 = 50 \text{ mg/l}$; $t_{Ec} = 5 \text{ min}$; $\text{pH} = 5.35$; $\gamma = 16.5 \text{ mS/cm}$; stirring speed = 200 rpm.

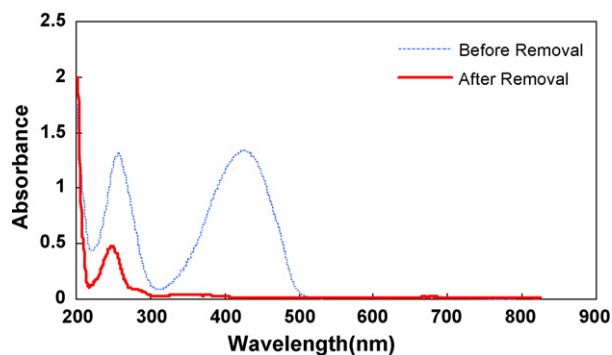


Fig. 10. Absorbance spectra of dye solutions containing C.I. Acid Yellow 23 before and after EC in the optimized conditions.

4.7. COD and absorbance spectra reduction of the dye solution

The COD of the dye solution was measured according to the standard methods for the examination of water and wastewater [26], and consequently decolorized in the optimized condition by iron electrodes: ($CD = 112.5 \text{ A/m}^2$, $C_0 = 50 \text{ mg/l}$, $t_{Ec} = 5 \text{ min}$, $\text{pH} = 5.35$, $\gamma = 16.5 \text{ mS/cm}$ and stirring speed = 200 rpm) and for dye solution containing C.I. Acid Yellow 23, the COD of the treated solution was measured again. The COD was reduced more than 69% ($\text{COD}_i = 123 \text{ mg/l O}_2$). Fig. 10 illustrates the adsorption spectra for dye solution before and after EC process using optimized experimental values. It can be seen that EC process in the optimized conditions caused almost complete color removal from the dye solution.

5. Conclusion

Electrocoagulation is one of the most effective techniques to remove color and organic pollutants from wastewater. The decolorization of dye solution, C.I. Acid Yellow 23, by electrocoagulation was affected by the current density (CD), time of electrolysis, initial pH, initial dye concentration and electrode material. Our results showed that the current density was the most effective parameter. In this work, it was shown that the electrocoagulation treatment results in effective and fast removal of C.I. Acid Yellow 23. For a solution with dye concen-

tration of 50 mg/l, color and COD elimination of 99% and 69%, respectively, when the pH was about 6, time of electrolysis was 5 min and current density was approximately 112.5 A/m^2 . Meanwhile, our experimental results showed that the cell voltage and SEEC for achieving the optimum current density increased with an increase in the interelectrode distance and a decrease in the conductivity and the area of cross-section of the electrodes.

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